Investigation for Magnetic Structure and Phase Boundary of (Sr,Ba)FeO3 with an Unusually High Valence State of Iron

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There have been known a small class of perovskite oxides that contain iron in an unusually high valence state of Fe4+ like SrFeO3 and CaFeO3. Their electronic properties are peculiar. Normal Fe2+- and Fe3+-oxides such as Fe2+O and LaFe3+O3 are antiferromagnetic insulators governed by the Kanamori-Goodenough rule, while these Fe4+-oxides exhibit a shift toward metallicity and ferromagnetism. SrFe4+O3 is a cubic metal in the proper screw spin structure with propagation vector along [111]. CaFe4+O3 shows a metal-to-insulator transition by charge-disproportionation to Fe3+ and Fe5+ at 290 K. It is notable here that Fe3+L (t2g3eg2L, S=2), i.e., a charge transfer from O to Fe, is the realistic picture in these oxides. The compounds with Fe4+ are thus quite attractive in terms of both experimental and theoretical views, but the knowledge of the Fe4+ physics is still far from satisfactory due to few model materials.

Until recently, perovskite BaFeO3 was only reported as thin film and the discussion about oxygen defects involves ambiguities and inconsistencies. However, three years ago, we reported BaFeO3 without any oxygen defect. BaFeO3 shows a proper screw spin structure along [100] having a different propagation vector from that of SrFeO3. In addition, we have found that this material exhibits a complete saturation magnetization (3.5 μ B) by the application of a tiny magnetic field (˜0.3 T). Thus, this material represents the first example to show ferromagnetism at ambient pressure. Subsequently, we have synthesized a powder sample of (Sr1-xBax)FeO3 ($0 < x < 1$) without any oxygen defect, and demonstrated magnetic phase diagram. We revealed a propagation vector [111] of (Sr1-xBax)FeO3 ($0 < x < 0.8$). Here, we want to emphasize is that the magnetic phase diagram in AFeO3 (A = Sr1-xBax) is very well consistent with that predicted by a theory demonstrated by Mostvoy. However, there still exist unrevealed magnetic phase, A-AFM, in between A-HM (magnetic structure of BaFeO3) and G-HM (magnetic structure of SrFeO3).

In order to explore this phase and reveal the phase boundary, we carried out neutron diffraction measurements of (Sr1-xBax)FeO3 ($0.8 < x < 1$) at Echinda of ANSTO. In addition, we also carried out neutron diffraction measurements of BaFeO3 under field.

We carried out neutron diffraction measurement of (Sr1-xBax)FeO3 (x = 0.2, 0.6, 0.8, 0.9, 1.0) at 10 K. Although satellite peaks can be observed in $0 < x < 0.8$, we could not observe any magnetic peaks for (Sr0.1Ba0.9)FeO3 and BaFeO3 because the resolution is not good enough.

We also carried out neutron diffraction measurement of BaFeO3 under 0 T and 2T at 3K. At 2T, the intensities for each peaks especially the peaks for lower angle increase, indicating that helical to ferromagnetic transition occurs. This is well corresponds to the phase diagram obtained by magnetic susceptibility measurement.

This experiment was performed by using Echinda at ANSTO, Australia, which was transferred from HERMES with the approval of Institute for Solid State Physics, The University of Tokyo (proposal no. 12700), Japan Atomic Energy Agency, Tokai, Japan.